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Dynamic response of a magnetic suspension in a viscoelastic fluid

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We present a model to describe a dynamic orientational response of a dilute suspension of single-domain magnetic particles in a viscoelastic liquid carrier of the Maxwell fluid type. Introducing Brownian orientational diffusion into the equation of the particle rotary motion, absorption spectra of the initial dynamic magnetic susceptibility are obtained. Under appropriate conditions they display a complex comblike structure which might be helpful for probing the local rheology of the carrier medium of a suspension. [S1063-651X(96)06609-3]

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I. INTRODUCTION

For a variety of fine magnetic particle dispersions in liquid matrices one may propose a conventional classification with regard to the fluidity features of the carrier. Namely, suspensions in Newtonian fluids, widely known as *magnetic fluids* or *ferrofluids*; suspensions in liquid crystals as well called *ferroliquid crystals*; and suspensions in non-Newtonian fluids.

Of these three kinds of fluid nanocomposites, the magnetic fluids have been extensively studied for a long time—see books [1,2] and bibliographies [3]. Ferroliquid crystals, initially addressed in Refs. [4,5], are now under progressive investigation [6,7]. However, the last item of the list has hardly been touched as yet.

Even after having been distinguished from ordinary and liquid-crystalline systems, ‘‘non-Newtonian magnetic suspensions’’ is probably too general a name to use. One just has to recall how variable in their properties possible carriers (polymeric melt or solutions, biological systems, etc.) may be.

Magnetic response of such media is to a great extent determined by the mobility properties of the particles in their fluid environment. For diluted suspensions, assuming that the embedded particles are magnetically hard, the most essential process is the orientational motion of an individual particle inside the non-Newtonian matrix.

This paper carries on the theoretical studies of kinetics of suspensions in viscoelastic media set out in Refs. [8–10]. We derive and analyze the dynamic susceptibility of a statistical assembly of fine ferroparticles in a Maxwell fluid. The latter is a simple conventional model of a polymeric solution. Of course, due to its simplicity, it gives up a lot of details relevant for the behavior of a particular polymer. The gain, however, is the opportunity to get acquainted with the effect of retardation—probably the most general non-Newtonian feature of polymeric solutions—on the frequency-dependent magnetic response.

The applicational aspect of the present and similar [11] studies is easy to recognize. Imagine a tiny amount of fine dipolar particles dispersed over the volume of a tested medium. Their motion is excited contactlessly by means of an alternating external field that does not affect the liquid matrix, and the dynamic polarization is recorded. Provided an adequate interpretation is available, the amplitude-frequency dependences of the response signal can yield essential information on the local rheology of the carrier fluid. A successful attempt of such an approach has been reported in Ref. [12]. Apparently, more theoretical effort is needed for further progress.

II. MAGNETIC PARTICLE IN A VISCOELASTIC FLUID

Let us consider a two-dimensional (2D) orientational motion of a single-domain magnetically rigid particle in a viscoelastic fluid. We choose the 2D representation for its utter simplicity. The 3D one, though far more cumbersome in mathematical treatment, would provide results differing from ours by but numerical coefficients of the order of unity.

In the adopted framework a magnetic particle is a solid disk of radius a confined in an arbitrary plane passing through the direction of the applied magnetic field $\mathbf{H}$, which we take for the polar axis of the coordinate system. The magnetic moment $\boldsymbol{\mu}$ of the particle is constant in its absolute value and its direction inside the plane of the disk is fixed. Orientation of the vector $\boldsymbol{\mu}$ with respect to the polar axis is defined by the angle ϑ. Then the equation of the particle rotary motion is written

$$I\ddot{\vartheta}+Q(t)+\mu H\sin\vartheta=y(t). \tag{1}$$

Here the dots denote differentiation with respect to time, I is the moment of inertia of a particle when rotated about its center, $Q(t)$ is the resistance torque, and the noise torque $y(t)$ accounts for thermal motion in the fluid.

Adopting the Maxwell model of a viscoelastic fluid we assume that the resistance torque exerted by the fluid carrier on a rotating particle may be described by the equation

$$\dot{Q}=-\frac{1}{\tau_M}(Q-Q_0),\quad \text{with}\ \ Q_0=\zeta\dot{\vartheta}, \tag{2}$$

where ζ is the friction coefficient proportional to the fluid viscosity η, and τ_M is the characteristic time of the elastic stress relaxation. Both η and τ_M are the characteristics of the fluid proper and as such are insensitive to the presence of particles.

Though any extensive discussion of the Maxwell rheological model is beyond the scope of the present work, it seems reasonable to recall that relationship (2) may be regarded as a valid one only for the fluids whose viscoelasticity is to some extent pronounced, viz., $\tau_M \gg \tau_I$, with the limit $\tau_M/\tau_I \lesssim 1$ being possibly incorrect. Here $\tau_I \sim I/\zeta$ is the inertial relaxation time—see Eq. (10) below. Indeed, for a consistent derivation of the set of local equations (1) and (2) a conjugated problem of field-induced rotations of a solid body and invoked by that velocity and deformation distributions in the surrounding viscoelastic fluid should be solved. Among other effects, for $\tau_M \lesssim \tau_I$ at high frequencies a skin-layer behavior must be expected with its specific transverse-wave dispersion law $Q \sim (i-1)\sqrt{\omega}$, well known for Newtonian fluids and not at all reflected in Eq. (2). As a partial justification of our use of Eq. (2), we remark that for the Reynolds numbers $\mathrm{Re} \leqslant 1$ this type of mechanical response takes place when the skin-layer thickness $\delta \sim \sqrt{\eta/\rho\omega}$ becomes comparable with the particle size. However, for rotary oscillations the conditions $\mathrm{Re} \leqslant 1$ and $a < \delta$ reduce to one: $\omega\tau_I < 1$, which in fact is adopted throughout our consideration.

To clarify the meaning of the relaxational equation (2), let us substitute there the angular displacement in the form $\vartheta = \vartheta^{(0)}\exp(-i\omega t)$. For the torque amplitude one finds

$$\mathrm{Re}Q^{(0)} = \frac{\zeta\omega^2\tau_M}{1+\omega^2\tau_M^2}\,\vartheta^{(0)}, \qquad \mathrm{Im}Q^{(0)} = \frac{\zeta\omega}{1+\omega^2\tau_M^2}\,\vartheta^{(0)}.$$

At low frequencies $(\omega\tau_M \ll 1)$ only the imaginary part of the resistance is relevant, which leads to $Q \approx -i\,\mathrm{Im}Q \propto \dot{\vartheta}$, meaning that the torque is predominantly viscous. The case of the Newtonian fluid is recovered at $\omega \to 0$, where $Q = Q_0$—see Eq. (2). At high frequencies one has $Q \approx \mathrm{Re}Q \propto \vartheta$, as if the medium were transformed into a Hookean elastic body with the effective strain modulus η/τ_M. In this case the response has a reversible, nondissipative, character. In the intermediate range the reaction of the fluid is a superposition of viscous (fluidlike) and elastic (solidlike) contributions.

As has been shown in Ref. [9], the random torque in Eq. (1) is ‘‘colored’’ with a correlation time τ_M. However, introducing an auxiliary white noise $f(t)$, one may treat $y(t)$ as a dynamic variable which evolves according to the equation

$$\hat{R}y \equiv \left(1+\tau_M\frac{d}{dt}\right)y = f(t), \qquad \langle f(t)f(0)\rangle = 2\zeta k_BT\delta(t). \tag{3}$$

Adding these relationships to Eqs. (1) and (2), one obtains a closed set of stochastic equations with a standard noise. Note that the linear retardation operator $\hat{R}$ entering the left-hand side of Eq. (3) coincides with that of Eq. (2) for Q. That means that the thermal and resistance torques have the same type of frequency dispersion. Upon elimination of Q and y, Eqs. (1)–(3) unite, rendering the Langevin equation of the problem that reads

$$\dddot{\vartheta} + \frac{1}{\tau_M}\ddot{\vartheta} + \frac{1}{I}\left(\frac{\zeta}{\tau_M} + \mu H\cos\vartheta\right)\dot{\vartheta} = -\frac{\mu}{I\tau_M}(\sin\vartheta)\hat{R}H + \frac{1}{I\tau_M}f(t). \tag{4}$$

Equation (4) provides a starting point for any statistical description of the particle assembly in question, derivation of the Fokker-Planck equation, for example.

III. DYNAMIC SUSCEPTIBILITY

Some results on formulation and solution of the corresponding kinetic equation are presented elsewhere [10,13]. This way is rather universal but laborious as well. To obtain the initial susceptibility, which is our main goal here, it is more feasible to use the linear response approach. According to the Kubo-Tomita theorem (see, [14] for example), the dynamic susceptibility may be expressed through the equilibrium correlation function of the observable quantity. For the system in question we choose as such the component of the induced magnetization in the direction of the probing field **H**:

$$M(t) = c\mu p(t), \quad \text{where } p(t) = p[\vartheta(t)] \equiv \cos\vartheta(t),$$

and c is the number concentration of the particles. Then the pertinent formula reads

$$\chi(\omega)/\chi_0 = -\int_0^\infty dt\,e^{i\omega t}\,\frac{d}{dt}\frac{\langle p(t)p(0)\rangle}{\langle p^2\rangle}, \tag{5}$$

where $\chi_0 = c\mu^2/2k_BT$ is the static susceptibility of an assembly of noninteracting magnetic dipoles, and the angular brackets denote the statistical average over the equilibrium field-free $(\mathbf{H}=0)$ state.

Using the well-known trigonometry formulas, the correlation function may be rearranged as

$$\langle p(t)p(0)\rangle = \tfrac{1}{2}\langle p[\vartheta(t)-\vartheta(0)] + p[\vartheta(t)+\vartheta(0)]\rangle = \tfrac{1}{2}\langle p(\Delta\vartheta)\rangle. \tag{6}$$

Here $\Delta\vartheta = \vartheta(t) - \vartheta(0)$, and for a stationary process the term containing the sum of angles vanishes. For evaluation of the equilibrium correlation function, one has to know the statistical properties of the assembly. It is easy to see that the angle ϑ is the Gaussian random variable. Indeed, according to Eq. (4), at $\mathbf{H}=0$ it obeys a linear equation, the right-hand side of which is the Gaussian function—the white noise f. Performing the averaging with the Gaussian distribution function, one gets (see Ref. [15], for example)

$$\langle p(\Delta\vartheta)\rangle = \exp[-\tfrac{1}{2}\langle(\Delta\vartheta)^2\rangle], \tag{7}$$

thus expressing the sought for dipolar correlation function via the mean-square angular fluctuation.

To find the latter, let us present the field-free solution of Eq. (4) as

$$\dot{\vartheta}(t)=\frac{\dot{\vartheta}(0)}{\Delta\lambda}(\lambda_+e^{\lambda_-t}-\lambda_-e^{\lambda_+t})+\frac{\ddot{\vartheta}(0)}{\Delta\lambda}(e^{\lambda_+t}-e^{\lambda_-t})$$
$$+\frac{1}{I\tau_M\Delta\lambda}\int_0^t dt'f(t')[e^{\lambda_+(t-t')}-e^{\lambda_-(t-t')}],\quad (8)$$

where $\dot{\vartheta}(0)$ and $\ddot{\vartheta}(0)$ are the initial values of the angular velocity and acceleration, respectively, and the decrements λ are defined by the characteristic equation

$$\lambda^2+\lambda/\tau_M+1/\tau_M\tau_I=0.\quad (9)$$

Here we have introduced the ''inertial'' time

$$\tau_I=I/\zeta,\quad (10)$$

determining the relaxation rate of the angular momentum for a particle with the moment of inertia I and friction coefficient ζ. The solution of Eq. (9) yields

$$\lambda_\pm=-\frac{1}{2\tau_M}\pm i\sqrt{\frac{1}{\tau_I\tau_M}-\frac{1}{4\tau_M^2}},\quad \Delta\lambda=\lambda_+-\lambda_-.$$

Therefore one sees that at $\tau_M\gg\tau_I/4$ the system behaves as an oscillator with eigenfrequencies resulting from the interplay between the inertia of the particle and the elasticity of its environment. Multiplying Eq. (8) by $\dot{\vartheta}(0)$ and averaging over the assembly, one finds

$$\langle\dot{\vartheta}(t)\dot{\vartheta}(0)\rangle=\langle\dot{\vartheta}^2(0)\rangle[\lambda_+e^{\lambda_-t}-\lambda_-e^{\lambda_+t}]/\Delta\lambda.\quad (11)$$

While writing down this expression, we have taken into account that $\langle\dot{\vartheta}(0)\ddot{\vartheta}(0)\rangle=0$.

Since the phase and the angular velocity are related by definition

$$\Delta\vartheta(t)=\int_0^t dt'\,\dot{\vartheta}(t'),$$

the general expression for the equilibrium mean-square fluctuation of the phase is

$$\langle(\Delta\vartheta)^2\rangle=2\int_0^t dt'\int_0^{t'}dt''\langle\dot{\vartheta}(0)\dot{\vartheta}(t'-t'')\rangle.\quad (12)$$

Substitution of Eq. (11) in (12) upon integration gives

$$\langle(\Delta\vartheta)^2\rangle=\frac{2k_BT}{I\Delta\lambda}\left[\frac{\lambda_-^2-\lambda_+^2}{\lambda_+\lambda_-}t+\frac{\lambda_-^3-\lambda_+^3}{\lambda_+^2\lambda_-^2}+\frac{\lambda_+}{\lambda_-^2}e^{\lambda_-t}-\frac{\lambda_-}{\lambda_+^2}e^{\lambda_+t}\right],$$

where we assume equipartition for the mean-square angular velocity, i.e., $\langle\dot{\vartheta}^2(0)\rangle=k_BT/I$. After simple algebraic transformations the mean fluctuation takes the form

$$\langle(\Delta\vartheta)^2\rangle=\frac{2}{\tau_D}\left[t+\tau_M-\tau_I+\frac{\tau_M}{\sqrt{1-1/4M}}\times\exp\left(-\frac{t}{2\tau_M}\right)\cos(\omega_Mt-\psi)\right],\quad (13)$$

with

$$\omega_M=\frac{1}{2\tau_M}\sqrt{4M-1},\quad \tan\psi=\frac{1-3M}{(1-M)\sqrt{4M-1}},$$
$$M=\frac{\tau_M}{\tau_I}.\quad (14)$$

As we are dealing with a thermalized system, the rate of the particle orientational diffusion determined by the Debye time

$$\tau_D=\zeta/k_BT\quad (15)$$

necessarily enters Eq. (13). For extremely short times ($\lambda t\ll1$) Eq. (13) yields the dynamic result

$$\langle(\Delta\vartheta)^2\rangle=(k_BT/I)t^2=\langle\dot{\vartheta}^2\rangle t^2,$$

and in the Newtonian fluid limit ($\tau_M\to0$) it reduces to the well-known relationship (see, [15] for example)

$$\langle(\Delta\vartheta)^2\rangle=\frac{2}{\tau_D}[t-\tau_I(1-e^{-t/\tau_I})].$$

Substituting sequentially Eq. (13) into (6) and (7) and those into Eq. (5), one gets for the initial dynamic susceptibility of a viscoelastic magnetic suspension

$$\chi(\omega)/\chi_0=1+i\omega\int_0^\infty dte^{i\omega t}G(t),\quad (16)$$

$$G(t)=\exp\left\{\frac{1-M}{D}\left[1-e^{-t/2\tau_M}\frac{\cos(\omega_Mt-\psi)}{\cos\psi}\right]\right\}e^{-t/\tau_D},\quad (17)$$

where $D=\tau_D/\tau_I$. From formula (17) it is clear that the relaxation time ratios M and D are the principal dimensionless parameters of the problem.

It is important to emphasize that the correlator (17) for the observable quantity, i.e., magnetization, unlike the angular correlator (13), is a nonlinear function of the phase fluctuation. Due to that, as it is shown below, the spectrum of the dipolar susceptibility may contain multiples of the unique resonance frequency inherent to more simple, but essentially nonobservable, angle oscillations. The same circumstance accounts for the additivity of orientational diffusion and viscoelasticity contributions to Eq. (13) and multiplicativity of those in Eq. (17).

Even an approximate analysis of the obtained susceptibility (16) and (17) is rather cumbersome. It is due, in particular, to a number of reference times in Eqs. (13) and (17). Indeed, besides three relaxation times, viz., τ_I, τ_M, and τ_D, one more time scale, namely, the eigenfrequency $\omega_M=1/\sqrt{\tau_I\tau_M}$ of particle rotary oscillations defined by Eq. (14), enters the pertinent formulas. To facilitate considerations, let us take the inertial time τ_I as a minimal time scale. This choice is ensured by the fact that its characteristic value even for rather large grains ($a\sim10^3$ nm) and inviscid liquids ($\eta\sim10^{-2}$ P, like water) is rather small ($\tau_I\sim a^2/10\eta\sim10^{-6}$ s). With an objective to study the effect of viscoelasticity, we assume that

$$\tau_I \ll \sqrt{\tau_M \tau_I} \ll \tau_M, \tag{18}$$

see also the comment following Eq. (2). Given that, one may expect that the actual form of the frequency dependence of the dynamic susceptibility is determined by the relative position of the Debye time τ_D in the time scale (18). Therefore at least three kinds of suspensions with regard to the matrix viscoelasticity (VE) must be distinguished:

$$\begin{aligned} &\sqrt{\tau_M \tau_I} < \tau_M < \tau_D \quad \text{(weak)},\\ &\sqrt{\tau_M \tau_I} < \tau_D < \tau_M \quad \text{(developed)},\\ &\tau_D < \sqrt{\tau_M \tau_I} < \tau_M \quad \text{(strong)}. \end{aligned} \tag{19}$$

The essential differences between the spectra of the systems with different VE extent may be seen right from Fig. 1 (quantitative evaluations are given in the following section), where curve *a* corresponds to weak, *c* to developed, and *d* to strong VE, whereas curve *b* is the border case between weak and developed VE.

IV. ABSORPTION SPECTRA

The integral Eq. (16) cannot be taken analytically. However, its numerical evaluation does not cause too many difficulties. A set of numerically obtained curves for the absorption lines $\chi'' = \mathrm{Im}\chi(\omega)$ is presented in Fig. 1. To improve their understanding, let us first establish some general properties of the dipolar susceptibility. To do that, we expand the correlation function (17) in a series with respect to the dimensionless parameter $\tau_M/\tau_D = M/D$ under the assumption that both $M, D \gg 1$. This yields

$$G(t) = e^{-q} \sum_{k=0}^{\infty} \frac{q^k}{k!(\cos\psi)^k} \exp(-\gamma_k t) \cos^k(\omega_M t - \psi), \tag{20}$$

where

$$q = \frac{\tau_M}{\tau_D}, \quad \gamma_k = \frac{1}{\tau_D} + \frac{k}{2\tau_M}, \quad \omega_M = \frac{1}{\sqrt{\tau_M \tau_I}}, \quad \tan\psi = \frac{3}{2\sqrt{M}}. \tag{21}$$

Replacing cosine by its complex representation, one arrives at

$$G(t) = e^{-q} \sum_{k=0}^{\infty} \frac{(\tilde{q}/2)^k}{k!} \exp(-\gamma_k t) \sum_{l=0}^{k} C_l^k \exp[i(k-2l)\alpha]. \tag{22}$$

Here $\tilde{q} = q/\cos\psi$, $\alpha = \omega_M t - \psi$, and $C_l^k = k!/(k-l)!l!$ is a binomial coefficient. The double sum (22) may be transformed into one in which the exponents equalling multiples of the eigenfrequency ω_M are ordered, that gives

$$G(t) = e^{-q} \sum_{n=0,\pm1,\pm2,\dots} e^{i\alpha n} \sum_{l=0}^{\infty} \frac{(\tilde{q}/2)^{|n|+2l}}{l!(|n|+l)!} e^{-\gamma_{|n|+2l} t}. \tag{23}$$

While writing down expansion (23) we have taken into account that its amplitudes are even in the index n. The same formula may be obtained directly from expansion (22) upon substitution $n = k - 2l$ and appropriate change of the summation limits.

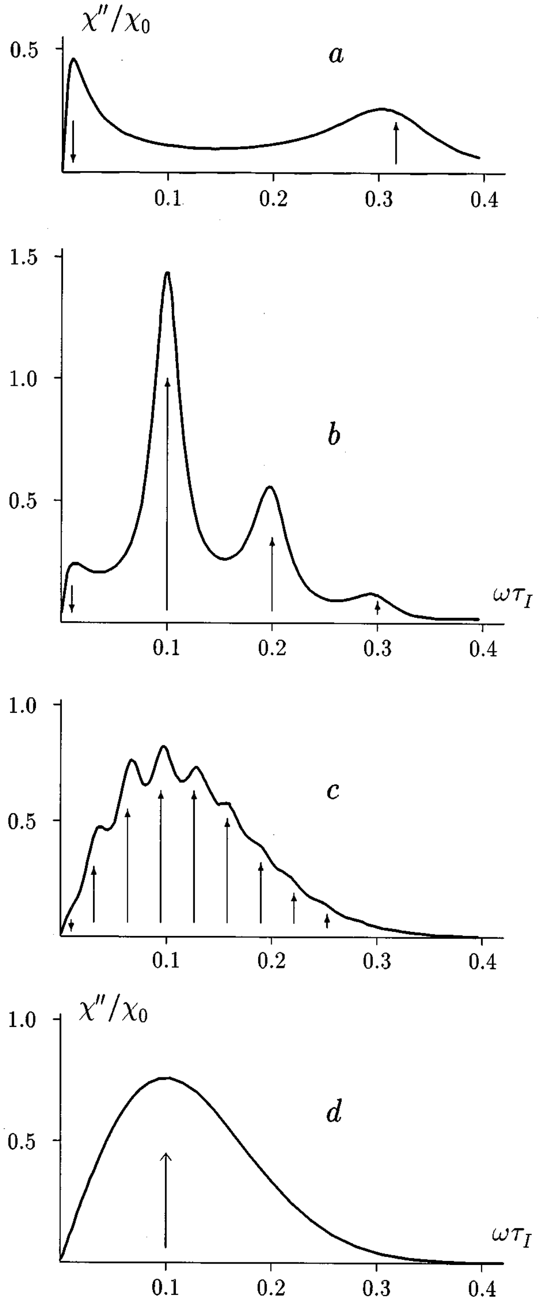

FIG. 1. Absorption lines for a situation when the particle size is constant ($D = 10^2$) whereas the stress relaxation time of the carrier fluid increases: curves *a*–*d* correspond to $M = 10$ ($q = 0.1$), 10^2 ($q = 1$), 10^3 ($q = 10$), and 10^4 ($q = 10^2$). In the framework of conditions (19), case *a* accounts for weak, *b* and *c* for developed, and *d* for strong viscoelasticity of a suspension. In patterns *a*–*c* downward arrows show the position where the Debye relaxational maximum should be located; upward arrows indicate the points $\omega = \omega_M, 2\omega_M, \dots$ of approximate locations of the maxima of the comblike spectra. The arrow in curve *d* corresponds to the position of the mean thermal angular velocity.

Using correlator (23) in the Kubo formula (16), and performing the now elementary time integration, one finally gets the representation of the susceptibility as

$$\chi(\omega)/\chi_0=1+i\omega e^{-q}\sum_{l=0}^{\infty}\frac{(\tilde{q}/2)^{2l}}{l!}\left\{\frac{1}{(\gamma_{2l}-i\omega)l!}+2\sum_{n=1}^{\infty}\frac{(\tilde{q}/2)^n}{(n+l)!}\,\frac{(\gamma_{n+2l}-i\omega)\cos(n\psi)+n\omega_M\sin(n\psi)}{(n\omega_M)^2+\gamma_{n+2l}^2-\omega^2-2i\omega\gamma_{n+2l}}\right\}. \tag{24}$$

The latter expression is convenient for a qualitative analysis. Let us begin with the case of weak VE, i.e., $\tau_M \ll \tau_D$ as defined by Eq. (19) or $q \ll 1$ in Eq. (24). For that with the first-order accuracy one finds

$$\chi(\omega)/\chi_0=\frac{1-q}{1-i\omega\tau_D}+\frac{q\omega_M}{2(\omega_M-\omega)-i/\tau_M}, \quad q\ll 1. \tag{25}$$

We remark that this result coincides with the one obtained in Ref. [9] by a direct solution of the Langevin equation (4) using the approximate method by Coffey [16].

The meaning of Eq. (25) is clear. Its first term yields the usual Debye susceptibility but slightly reduced by a weak viscoelasticity of the carrier liquid. The second term, being a Lorentzian, reflects the oscillatory character of relaxation in an assembly of magnetic particles embedded in a medium possessing dynamic elasticity. All the overtones ($2\omega_M$ and higher) in this limit are beyond resolution. When transformed to the absorption spectrum $\chi''(\omega)$, formula (25) gives curves very close to that presented in curve *a* of Fig. 1. As Eq. (25) shows, the width of the right peak (VE resonance) of the imaginary part of the susceptibility is $\Delta\omega \approx 1/\tau_M$ and its height is $\chi''_{\max} \sim \tau_M\sqrt{M}/\tau_D$. This means that the peak becomes untraceable as soon as $\tau_M\sqrt{M} \ll \tau_D$. Therefore the latter relationship may be regarded as the ultimate observability condition for a viscoelasticity of magnetic suspensions, below which from the magnetic spectroscopy viewpoint they do not differ from ordinary Newtonian ones.

As the viscoelasticity increases (parameter q grows and exceeds unity), the case of developed VE [see conditions (19)] takes place. Then, according to formula (24), the peaks at multiple ($2\omega_M$, $3\omega_M$, ...) frequencies become pronounced. In the vicinity of each resonance, expression (24) simplifies considerably, and the susceptibility may be presented as a superposition of Lorentzians:

$$\chi(\omega)/\chi_0 \simeq 1+e^{-q}\sum_{n=1}^{\infty}\Omega_n\sum_{l=0}^{\infty}\frac{(q/2)^{n+2l}}{(n+l)!\,l!}\,\frac{(\Omega_n-\omega)+i\gamma_{n+2l}}{(\Omega_n-\omega)^2+\gamma_{n+2l}^2},$$

$$|\Omega_n-\omega| \ll \omega_M \tag{26}$$

where $\Omega_n=\sqrt{(n\omega_M)^2+\gamma_{n+2l}^2} \simeq n\omega_M$. The imaginary part of the susceptibility, i.e., the function proportional to absorption, emerges from (26) as a sequence of equidistant peaks (a comblike pattern). This kind of behavior is illustrated by curves *b* and *c* of Figs. 1. The numerical analysis shows that the curves enveloping the combs descend monotonically when the ratio of the relaxation times $q=\tau_M/\tau_D<2$, whereas for $q>2$ they have a maximum which lies approximately at the frequency $\omega_M\sqrt{q}$. We remark that the latter quantity is equal to the mean value of the particle angular velocity $\overline{\omega}=\sqrt{k_BT/I}$ over the Gaussian distribution. As such it is a fundamental characteristic of the equilibrium statistical assembly, and does not depend on the rheological parameters of the carrier liquid.

Together with the enveloping profile, the viscoelasticity growth affects the structure of each peak. At $q<2$ the resolved peaks with high accuracy are single Lorentzians homogeneously broadened. But at $q>2$ each peak turns out to be a pack of closely positioned Lorentzians with comparable amplitudes that means domination of the nonhomogeneous broadening. Unlike weak VE, in a system with a developed viscoelasticity the rate of particle rotary diffusion is equal to or higher than that of stress relaxation. Under the joint influence of both processes the effective width of a peak becomes

$$\Delta\omega_n \approx \gamma_n=\frac{1}{\tau_D}+\frac{n}{2\tau_M}$$

[see Eq. (21)] where now both terms are relevant.

On further increase of q, the number of resolved peaks grows unboundedly ($\sim\sqrt{q}$) whereas their widths remain virtually constant $\Delta\omega_n \simeq 1/\tau_D(1+n/q) \simeq 1/\tau_D$, where n is the peak's number. Simultaneously, the interpeak distances decrease as $\omega_M=\overline{\omega}/\sqrt{q}$. This ends up at the limiting contour where the peaks eventually fuse, forming a smooth cusp whose maximum sits at $\overline{\omega}$—see curve *d* of Fig. 1. It is but natural that the condition of fusion of the neighboring peaks ($\omega_M\tau_D \lesssim 1$) renders the last line of Eq. (19), i.e., the strong VE limit.

Let us look at strong VE in more detail. The condition of peak fusion $\omega_M\tau_D \lesssim 1$ means that, unlike the cases of weak and developed VE, now the frequency of particle rotary oscillations becomes smaller than the rate of orientational (Debye) relaxation. In other words, these oscillations would not contribute to the susceptibility. For quantitative consideration, the previously used expansion with respect to parameter $q=\tau_M/\tau_D$ is not feasible any longer. However, revisiting the general expression (17) for the dipolar correlation function, it may be shown that for the system in question it reduces to

$$G(t)=\exp(-\overline{\omega}^2t^2/2), \quad \omega_M\tau_D<1, \tag{27}$$

reflecting the fact that under strong VE the main contribution to the susceptibility is yielded by the dynamical range of the phase fluctuation. During those short intervals one may assume that each particle of the assembly undergoes free rotation whereas the spreadout of the angular velocities is determined by the equilibrium distribution function. Hence $\langle\Delta\vartheta^2\rangle=\langle\dot{\vartheta}^2\rangle t^2=(k_BT/I)t^2$. Substitution of correlator (27) into the Kubo formula (16) gives

$$\chi(\omega)/\chi_0=1+i\omega\int_0^\infty dt\exp[i\omega t-(k_BT/2I)t^2]$$

$$=1-2x\exp(-x^2)\int_0^x dte^{t^2}+i\sqrt{\pi}xe^{-x^2}, \quad (28)$$

where $x=\omega/(\sqrt{2}\overline{\omega})$. The imaginary part has a maximum $\chi''_{\max}/\chi_0=\sqrt{\pi/2e}\approx 0.76$ at $x=\sqrt{2}/2$.

Therefore at strong viscoelasticity the dipolar susceptibility of a suspension tends to that of a system of noninteracting plane rotators (see Ref. [17], for example). As it might have been expected, in this limit the susceptibility lines are smooth with a maximum around $\overline{\omega}$, as in curve *d* of Fig. 1. The χ'' curve being nonsingular and positive means that the system absorbs the field energy despite vanishing of the contribution of the viscous term. For a qualitative understanding of such a behavior, one has to recall that in a thermalized assembly a number of particles which oscillate with the rates close to the external frequency ω is ever present. In fact, only this fraction of all the particles may indeed be considered to be in a strong coupling with the field. Of this number, those which have angular velocities slightly less than ω are accelerating and, thus, absorb power, whereas other particles decelerate, giving the power back to the field. Since in a thermodynamical (equilibrium) system the particle energy distribution $w(E)$ is always a diminishing function of E, dissipation always exceeds generation thus yielding positive absorption. This type of dissipation is known as *the Landau damping* [18]. In the molecular spectroscopy theory, where the formulation of the problem happens to be rather close to the one under discussion, the Landau damping has been considered in Ref. [17].

V. DISCUSSION

The presented results are the outcome of a kinetic treatment. To assess them more clearly, a comparison with some other model would have helped a lot. However, we do not know of any already developed model of that kind. Because of that we would propose a simple alternative by ourselves. A phenomenological concept is the first to cross one's mind when setting about a study of a viscoelastic suspension. The corresponding model might be called *the dispersive viscosity* approximation.

As is well known (see, [19] for example), the phenomenological equation governing the evolution of macroscopic magnetization in a suspension of rigid dipoles is

$$\tau_D\frac{dm}{dt}+m=\chi_0H, \quad (29)$$

where τ_D is defined as in Eq. (15). Inclusion of inertia transforms Eq. (29) into

$$\frac{I}{k_BT}\frac{d^2m}{dt^2}+\tau_D\frac{dm}{dt}+m=\chi_0H. \quad (30)$$

Now, bearing in mind that the resistance coefficient ζ and hence τ_D are proportional to the viscosity of the matrix, we introduce its dispersion. For that purpose we replace in Eq. (30) the resistance torque by some instantaneous value

$$\frac{I}{k_BT}\frac{d^2m}{dt^2}+\tilde{Q}+m=\chi_0H, \quad (31)$$

assuming that the stationary regime is achieved in a time period τ_M. Then the simplest equation determining $\tilde{Q}$ is written as

$$\tau_M\frac{d\tilde{Q}}{dt}=-\tilde{Q}+\tau_D\frac{dm}{dt}. \quad (32)$$

Equations (31) and (32) form a closed set from which $\tilde{Q}$ may be eliminated by differentiation to yield

$$\tau_I\tau_M\tau_D\frac{d^3m}{dt^3}+\tau_I\tau_D\frac{d^2m}{dt^2}+(\tau_D+\tau_M)\frac{dm}{dt}+m$$

$$=\chi_0\left(H+\tau_M\frac{dH}{dt}\right), \quad (33)$$

if we define the inertial time as in Sec. III. What is most essential here is that in the frame of the phenomenological approach Eq. (33), unlike Eq. (4), is considered to be macroscopic, i.e., final. The dynamic susceptibility yielded by Eq. (33) is

$$\frac{\chi}{\chi_0}=\frac{1-i\omega\tau_M}{(1-i\omega\tau_M)(1-\omega^2\tau_I\tau_D)-i\omega\tau_D}. \quad (34)$$

This expression coincides with our previous results [9,13] for χ obtained either by using the Coffey method or by a direct solution of the Fokker-Planck equation in the lowest order. It is easy to see that Eq. (34) always gives just one high-frequency peak of χ'', and never the comblike spectrum matching the weak VE case considered in Sec. IV. Therefore the dispersive viscosity assumption, however reasonable it may seem, is rather rough and has a limited validity. Its power is entirely exhausted at the point where the kinetic description just appeals for some extra accuracy. On doing that, the kinetic approach gives fruitful insight into the dynamic properties of the system and yields conclusions which are rather nontrivial.

We remark that certain results akin to those presented in Sec. II–IV have been dealt with some time ago in the molecular spectroscopy theory when linear non-Markovian stochastic processes were put under study. In particular, a multipeak spectral pattern has been obtained [20] with the itinerant oscillator approximation [21,15]. In this scheme a molecule is modeled by a conjunction of two coaxial rotators. The dipole moment is located in the inner one which is free except for being coupled through an elastic bond to the peripheral. This is the latter that is assumed to be subjected to both viscous resistance and random thermal torques. However, in molecular liquids the domain of material parameters where the multipeak structure might resolve turned out to be over the edge of the physical realm.

We also would like to point out a striking resemblance between the lines of the type of curve *c* of Fig. 1 and the far-infrared spectra of dipolar molecules embedded in nonpolar fluids. If we take a glance at the absorption lines of HCl dissolved in liquid noble gases which are analyzed in papers [22,23], both the series of equidistant spikes and the enveloping cusp centered at $\overline{\omega}$ are clearly distinguishable. However, one must be well aware that despite their topological coincidence those comblike patterns are of a completely different origin. Our curves are the results of a classic treatment whereas the mentioned molecular spectra reflect the quantum rotational structure of HCl. In particular, it is easy to see that the basic frequency for the pattern of curve *c* of Fig. 1 is $\omega_M = \sqrt{I\tau_M/\zeta}$ (and thus changing with the fluid viscoelasticity) but it is $\sim\hbar/I$ (and thus fundamental) for the HCl spectra.

Finally, let us show that with composite fluid media like viscoelastic magnetic suspensions, the multipeak patterns are well achievable, at least in principle. For estimates we take a spherical solid particle of a radius $a\sim 0.1\mu$m in a Maxwell fluid with the viscosity $\eta\sim 1$ P and the stress relaxation time $\tau_M\sim 0.1$s. Then it yields

$$\tau_I\sim a^2/\eta\sim 10^{-10}\ \text{s},\qquad \tau_D\sim a^3\eta/k_BT\sim 3\times 10^{-2}\ \text{s},$$

that corresponds to the dimensionless numbers $M=10^9$, $D=3\cdot 10^8$, the characteristic frequency $\omega_M\sim 3\cdot 10^5$ and the linewidth $\Delta\omega\sim 10-100\ \text{rad}^{-1}$, which are rather customary for magnetic susceptibility measurements. However, prior to an attempt of experimental verification of the given predictions, one essential point must be clarified; that is, how the distributions of stress relaxation times and particle sizes which are always present in any suspension would affect the fine structure of the spectral patterns.

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